

INTRODUCTORY COMMENTS

Claims 23 and 25 have been amended as indicated in the Amendments to the Claims. Claims 1-5, 7-23, 25-31, 36-64, 66, 67 and 69-71 are currently pending. Claims 1-5, 7-22, 43-46, 50-64, 66 and 67 have been withdrawn from consideration. Claims 6, 24, 32-35, 65 and 68 have been previously canceled. New Claim 72 has been added in this Amendment. Claims 23, 25-31, 36-42, 45-49, 65 and 69-72 remain for consideration by the Examiner. Please note that in the Amendments to the Claims, all pending, non-withdrawn claims are included herein for the convenience and efficiency of examination.

REMARKS

The Official Action dated July 16, 2008 in this Application has been carefully considered and the above amendments and following remarks are presented in a sincere attempt to place this Application in condition for allowance. Claims 23 and 25 have been amended and new Claim 75 has been added. Reconsideration and allowance are respectfully requested in light of the foregoing amendments and the following remarks.

By this Amendment, Claim 23 has been amended to clarify that the initial step is “forming a metal oxide or silicon oxide based sol-gel preformed material prior to reaction with an organic cross-linking agent,” then “contacting the preformed metal oxide or silicon oxide based sol-gel material with an organic cross-linking agent, the cross-linking agent comprising an organic compound that provides an organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material,” and then “drying the cross-linked metal oxide or silicon oxide based sol-gel preformed material provided with a conformal coating of chemically bound organic material to form the porous cross-linked metal oxide or silicon oxide based aerogel material.” (New language underlined.)

Claim 25 has been similarly amended to clarify that the cross-linked metal oxide or silicon oxide based sol-gel material comprises “[a] cross-linked metal oxide or silicon oxide based sol-gel material, comprising a metal oxide or silicon oxide based sol-gel preformed material that has been formed prior to reaction with an organic cross-linking agent,” and that the “metal oxide or silicon oxide based sol-gel preformed material [has] a conformal coating of an organic substance formed by chemical bonding of an organic cross-linking agent to surfaces of said metal oxide or silicon oxide based sol-gel preformed material after formation of said metal oxide or silicon oxide based sol-gel preformed material so as to form said cross-linked metal

oxide or silicon oxide based sol-gel material” (New language underlined.)

New Claim 72 is similar to Claim 25, but with an additional limitation specifying that “the volume relaxation energy (VRE) of the cross-linked metal oxide or silicon oxide based sol-gel material upon evaporation of the pore-filling solvent is less than the energy required to destroy the chemical bonds associated with cross-linking.” This additional limitation is supported in the original specification at, for example, page 14, line 10 to page 15, line 2).

Rejection of Claims 23, 25-31, 36-42, 47-49, 69, 70 and 71 under 35 U.S.C. 112, First Paragraph

The Examiner has rejected Claims 23, 25-31, 36-42, 47-49, 69, 70 and 71 under 35 U.S.C. 112, first paragraph, on the asserted basis that the claims fail to comply with the written description requirement. The Examiner contends that Applicants’ prior amendment inserting “connecting surface active groups of the metal oxide or silicon oxide based sol-gel material” into the claims lacks support in the originally filed disclosure. The Examiner does not agree that this language is supported by the specification at page 10, line 30 to page 11, line 4.

The foregoing rejection is respectfully traversed, and submitted to be inapplicable to the claims as currently amended. In particular, the original specifications, page 10, lines 12-27 [Par. 0057 of the Published Application], states that a diisocyanate cross linker is introduced into a preformed silica aerogel structure by taking hydrogels, which have been pre-prepared according to references by Leventis et al. Chem. Mater. 1999, 11, 2837-2845 and Leventis et al. NanoLetters 2002, 2, 63-67, and according to a post-gelation doping protocol (described in detail in the examples), they are filled with a diisocyanate solution by washing successively with methanol, propylene carbonate (PC) and PC/diisocyanate. The vials containing the gels in the last bath were heat-treated, cooled etc. and dried supercritically. Subsequently, in page 17, line 21 to page 18, line 11 [Pars. 0081-0083 of the Published Application] the specifications state:

EXAMPLES

Example I

[0081] • Preparation of Diisocyanate Cross-Linked Tetramethoxysilane Sol-Gel Monoliths

[0082] The contents of two vials: Vial A: 4.514 mL of tetramethoxysilane; 3.839 mL of methanol; and Vial B: 4.514 mL of methanol; 1.514 mL water and 20 μ L (microliter) of concentrated ammonium hydroxide, were mixed thoroughly to form the sol, which was poured in polyethylene cylindrical vials (1 cm diameter, 5 cm long). The molds were covered with parafilm®. To facilitate removal of the gels from the polyethylene molds, the bottom cup was covered with a teflon-tape lining. The sol gels in the molds at room temperature in about 10 min, and the resulting gels are left in the molds to age (i.e., complete polymerization and cross-linking) for 2 days. Subsequently, the bottom covers of the molds were removed together with the teflon lining, and the gels were pushed gently out of their molds directly into vials filled with methanol (one gel per vial).

[0083] The pores of the gel was then filled with a diisocyanate solution by washing the gel four times for eight hours per wash with methanol, propylene carbonate, and then a mixture containing propylene carbonate and poly(hexamethylene diisocyanate) in succession. During the last wash, the gels were heated to 100° C. for three days, and were then cooled to room temperature. The wash solution was decanted and the gels were washed with propylene carbonate one time for eight hours. The gels were then washed in a mixture of propylene carbonate and acetone (1:1, v/v) one time for eight hours, followed by washing in a mixture of propylene carbonate and acetone (1:3, v/v) one time for eight hours. The gels were then washed in acetone four times for eight hours per wash. Following the acetone wash, the gels were dried supercritically with carbon dioxide.

This description of [Pars. 0082-3] explicitly describes how to make the silica sol-gel preformed materials, prior to substantial cross-linking by organic cross-linking compounds (which does not occur, because organic cross-linking agents have not yet been added). Even specific details of how to manipulate the molds so that the preformed silica sol gels materials do not crack and so that they come out as perfect monoliths are also provided. Details on the concentrations of the isocyanate cross-linker in the cross-linking bath are given in Table II, page 7.

To avoid confusion, it should be noted that in the description above, the statement: “[R]esulting gels are left in the molds to age (i.e. complete polymerization and cross-linking)”

does not refer to cross-linking by *organic* cross-linking compounds. In this art, it is understood that the terms “polymerization and cross-linking” are used to describe the gelation and aging of a metal oxide or silicon dioxide sol gel material formed from its precursors, solvents and catalysts (for example, as in Example I above, tetramethosysilane, methanol and ammonium hydroxide, respectively), which is the formation of the metal oxide or silicon oxide particles themselves via chemical reactions leading to metal-oxygen-metal and silicon-oxygen-silicon bonds, and to formation of the network of particles via chemical reaction of the surface groups of the newly formed particles to form interparticle metal-oxygen-metal or Si-O-Si bridges. Those processes are known in the art and, though described in the present specification as “polymerization and as cross-linking,” respectively, clearly those terms do not refer to cross-linking by organic compounds. It is self-evident that cross-linking by organic compounds does not occur in the absence of organic cross-linking agents, and that such does not occur in the procedure of the present invention, where the cross-linking agents are added after the metal oxide or silicon oxide particle network has been preformed.

In the present invention, after formation of the sol-gel preformed material as described in [Par. 0082] in the subsequent step an organic cross-linker (which, in this example, is an isocyanate) is added, and chemically bonds with and conformally coats, the surfaces of the silicon oxide sol gel preformed material. *See*, [Par. 0069]. The spectroscopic analysis provided in page 5 of the Published Application under [Par. 0069] provides evidence that, in the present invention, the isocyanate cross-linker reacts and gets bonded chemically with active groups like silanols on the surface of the silica sol-gel preformed material:

IR analysis shows that as the density increases, the urethane C=O stretch (at $\sim 1690\text{ cm}^{-1}$) becomes comparable to, and eventually even stronger than, the Si—O stretch at 1078 cm^{-1} . Note also that while the urethane C=O stretch is present in di-ISO, the dominant stretch at $\sim 2272\text{ cm}^{-1}$ comes from the isocyanate (N=C=O) (Husing et al., Chem. Mater., 1998, 10, 3024-3032). However, the latter absorption is consistently extremely weak or absent. Therefore, it was concluded that both ends of practically all di-ISO have reacted. A typical SEM image of one of the most dense composites (FIG. 2B) shows that (a) a new material has been introduced conformally to the secondary particles, as not only the necklace-like structure but also individual particles remain clearly visible; and (b) the mesoporosity has been somewhat reduced, as several secondary particles appear fused (clustered) together, forming the larger domains that promote light scattering and haziness (Novak et al., Chem. Mater., 1994, 6, 282-286). These observations are all consistent with reaction and binding of di-ISO to the surface of silica. Considering the total surface area of native silica ($\sim 1000\text{ g cm}^{-3}$, Table II) and the density change between native silica and the most dense composite ($\rho_p = 0.447\text{ g cm}^{-3}$), it is calculated that the amount of di-ISO corresponds to 4.7 monolayers. Hence, terminal NCOs appear to undergo not only condensation with surface-silanol groups but also further reaction with themselves causing extensive cross-linking. It

The specification also discloses that metal oxide or silicon oxide (silica) based sol-gel materials are surface-terminated with hydroxyl groups (e.g., silanols). This is clearly stated in page 2 of the specifications under [Par. 0031]:

[0031] Silica is surface-terminated with silanols (—SiOH). A polyurethane, (—CONH—R—NHCOOR'—)_n, is formed by the reaction of a diisocyanate (OCN—R—NCO) and a diol (HO—R'—OH) (Smith and March, March's Advanced Organic Chemistry Reactions, Mechanism and Structure; John Wiley and Sons: New York, 2001). A similar reaction of an isocyanate cross-linking agent with the free hydroxyl groups of a sol-gel like material provides cross-linked aerogels having superior properties that allow them to be used in a large variety of applications.

In deference to the Examiner, however, and in order to expedite advancing this case to issue, Applicants have amended Claims 23 and 25 to delete the language that the Examiner contends is not sufficiently supported, and to replace it with new language specifying that the

preformed metal oxide or silicon oxide based sol-gel preformed material “ha[s]a conformal coating of an organic substance formed by chemical bonding of an organic cross-linking agent to surfaces of said metal oxide or silicon oxide based sol-gel preformed material after formation of said metal oxide or silicon oxide based sol-gel preformed material.” This language clearly finds explicit support in the [Par. 0069] of the specification, disclosing:

A typical SEM image of the one of the most dense composites (FIG. 2B) shows that (a) a new material has been introduced conformally to the secondary particles [of the silicon oxide sol gel perform], as not only the necklace-like structure but also individual particles remain clearly visible; and (b) the mesoporosity has been somewhat reduced, as several secondary particles appear fused (clustered) together. . . . These observations are all consistent with reaction and binding of di-ISO to the surface of silica . . . [and that] terminal NCOs appear to undergo not only condensation with surface-silanols but also further reaction with themselves causing extensive cross-linking . . .” Emphasis added).

In view of the foregoing amendments and remarks, it is respectfully submitted that the rejection of independent Claims 23 and 25 (and their respective dependent Claims 26-31, 36-42, 47-49, 69, 70 and 71) under 35 U.S.C. 112, first paragraph, is inapplicable to the Claims as amended and its withdrawal is courteously requested:

The examiner further rejects claims 26-31 on the asserted basis that the originally filed patent application “does not provide support of the range of amount values as claimed. It is not evident that the ranges of weight percent values now claimed are supported by the originally filed supporting disclosure for the products that are now claimed.”

This rejection is respectfully traversed. According to Claims 26-31 the amount of the cross-linking agent comprises between at least 2% and at least 80% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material. This is fully supported by the data in Table II (page 7), where it is disclosed that the density increases from 0.169 g/cc for the native (non-cross-linked) samples to 0.439 g/cc for the sample having 51% weight percentage di-ISO in the bath.

TABLE II

<u>Properties of Silica/Di-ISO Aerogel Monoliths</u>							
% di-ISO w/w in the bath	diameter ^b (cm)	ρ_b^b (g cm ⁻³)	BET area (m ² g ⁻¹) (Av.) pore diam. (nm)	hydrophilicity ^d	load at rupture ^e (kg)	diametral deflection at rupture ^f (cm)	modulus of elasticity (E) ^g (Mpa)
0.0	0.999 ± 0.002	0.169 ± 0.004	997 (13.4) ^c	66 ± 16	0.12 (h)	h	h
4.0	0.999 ± 0.002	0.241 ± 0.003	324 (i)	16 ± 2	1.75 (i)	0.061	5.3 (i)
8.0	0.986 ± 0.010	0.297 ± 0.007	308 (18.1)	8 ± 2	3.95 (4.38)	0.101	12.5 (19.5)
16	0.954 ± 0.011	0.388 ± 0.008	309 (22.3)	4 ± 1	9.65 (9.73)	0.140	i (20.4)
25	0.936 ± 0.007	0.390 ± 0.013	245 (21.9)	4 ± 1	10.4 (9.87)	0.129	31.3 (26.1)
34	0.908 ± 0.013	0.440 ± 0.009	215 (23.2)	5 ± 2	8.70 (9.87)	0.110	i (40.1)
42	0.907 ± 0.006	0.447 ± 0.020	165 (16.6)	5 ± 1	14.7 (15.6)	0.199	47.5 (46.3)
51	0.897 ± 0.014	0.439 ± 0.018	178 (29.8)	4 ± 2	12.0 (11.8)	0.109	i (53.8)

A person skilled in the art would understand that the weight percentage of the organic material formed by cross-linking can be readily calculated by taking into consideration the sample shrinkage as deduced from the sample diameters given in Table II, based on the formula:

$$\text{Organic material \% w/w} = \left[1 - \left[\left(\frac{\text{diameter}_x}{\text{diameter}_{\text{native}}} \right)^3 \left(\frac{\rho_x}{\rho_{\text{native}}} \right) \right]^{-1} \right] \times 100$$

For the case described above where 51% di-ISO weight percent in the bath is used (diameter of 0.897 cm and density of 0.439 gram/cm³, and it is found equal to 47% w/w.

Table I (page 7) describes an example having a different density:

TABLE I

<u>Properties of Supercritical Fluid Extraction (SFE) dried and Pentane dried aerogels</u>		
	Supercritical Fluid Dried	Pentane Dried
Density (g/cm ³)	0.57 ± 0.02	0.56 ± 0.01
BET Surface Area (m ² /g)	250 ± 10	132 ± 18
Average Pore Diameter (Å)	194 ± 8	185 ± 20

The same calculation applied to the gels described in Table I yields:

$$\text{Organic material \% w/w} = 100 \times (0.57 - 0.169)/0.57 = 70\% \text{ w/w.}$$

A person skilled in this art would readily understand that, to make material having a lesser amount of the cross-linking agent (i.e., the 2 percent amount specified in Claim 26), a

lesser percentage of di-ISO in the bath would be used. Such a person would also understand that, to make material having a greater amount of the cross-linking agent (i.e, the 80 percent amount specified in Claim 31), a greater percentage of percentage of di-ISO in the bath would be used. For the intermediate amounts specified in Claims 27-30, intermediate percentages of di-ISO in the bath would be used. Accordingly, the recitation of the ranges of amounts specified in Claims 26-31 are fully supported by the specification, and in such a way that it reasonably conveys to one skilled in the relevant art that the inventors, at the time the application was filed, had possession of the claimed invention.

In view of the foregoing amendments and remarks, it is respectfully submitted that the rejection of independent Claims 23 and 25 (and their respective dependent Claims 26-31 under 35 U.S.C. 112, first paragraph, is inapplicable to the Claims as amended and its withdrawal is courteously requested.

Rejection of Claims 1-24 Under 35 U.S.C. 102(b)

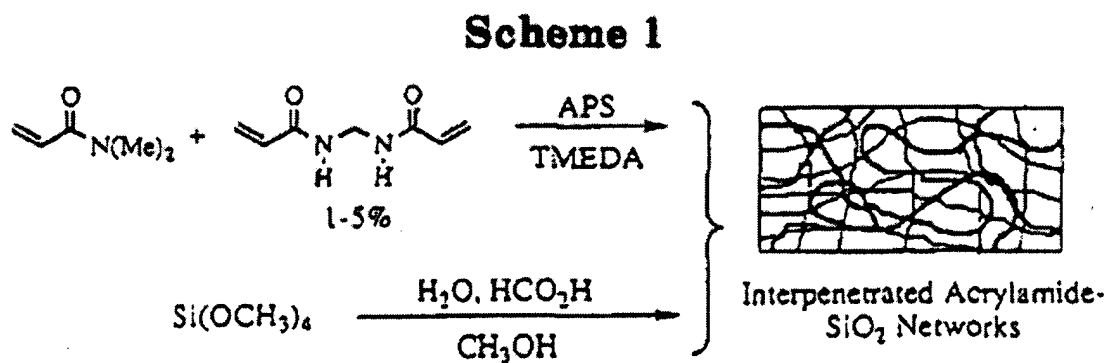
Turning to the substantive portions of the Official Action, the Examiner has rejected Claims 23, 25-31, 36, 39-42, 47-49, 69, 70 and 71 over under 35 U.S.C. 102(b) as assertedly being anticipated by Novak et al. Chem. Mater. 1994, 6, 282-286. The Examiner contends that "Novak et al. disclose preparations of crosslinked aerogels prepared by forming a sol-gel material as defined by applicants' claims, solvent to form a wet-gel films followed by drying...." The Examiner further contends that "applicants' arguments ... do not identify differences over Novak et al. which are supported by corresponding limitations in the claims."

To the extent the Examiner's rejection can apply to independent Claims 23 and 25 (and their respective dependent Claims), it is respectfully traversed.

Novak's materials, even according to the examiner's summary, are made by a two-step

process, the first of which involves simultaneously co-mixing silica sol-gel precursors along with either a polymer or in one instance with organic polymer-forming compounds. The second step involves drying the wet-gel.

Novak states that “under reaction conditions, the silanol end groups of the PDMS *cocondense* into the inorganic framework.” (Novak, at 283, col. 2, lines 19-21)(emphasis added). Novak teaches that his cocondensation approach results in a material having polymerized “strings” of polymer interpenetrating through the interstices of the silicon oxide network, as depicted in Novak’s “Scheme 1” figure. (Novak, page 284, col. 1, “Scheme 1”).



By contrast, in the present invention, a three step process is used. The first step of the present invention, which is absent in Novak, is “preforming a metal oxide or silicon oxide based sol-gel preformed material prior to reaction with organic cross-linking agents.” (Emphasis added.) Then, the preformed metal oxide or silicon oxide based sol-gel perform material is contacted with “an organic cross-linking agent, the cross-linking agent comprising an organic compound that provides an organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material.” The last step is “drying the cross-linked metal oxide or silicon oxide based sol-gel preformed material provided with a

conformal coating of chemically bound organic material to form the porous cross-linked metal oxide or silicon oxide based aerogel material.” (New language underlined.)

Accordingly, by contacting a cross-linking agent able to react chemically with the surface groups (e.g., silanols) of an already-formed metal oxide or silicon oxide preformed material, as in the present invention (rather than using cocodensation of the silicon oxide in the presence of the polymer, as in Novak), the present invention results in a material having “an organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material.” (Emphasis added.) Owing to this different method, Applicants’ resulting material is also different: a material having an “organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material” (emphasis added) rather than the “strings” of polymer interpenetrating through the interstices of the silicon oxide network, as depicted in Novak’s “Scheme 1” figure, that result from Novak’s method. (Novak, page 284, col. 1, “Scheme 1”).

Novak’s *interpenetrating organic networks* are totally different materials from ours. In our case we provide massive data in the specifications (see above) showing that the cross-linker reacts chemically with the surface of the silica nanoparticles and therefore it accumulates *conformally* on the inorganic nanoparticle network, as designed. Indeed, our SEMs (Figures 2A and 2B) do not show any foreign material (polymer) filling the mesopores, but instead, despite the massive cross-linker uptake (Applicants’ cross-linked samples contain more polymer than silica – see above), the material after cross-linking looks identical to the native material before cross-linking. Clearly, our material does differs from Novak’s not because of use of a different amount of a filler, but because of deposition of a conformal coating of polymer onto a preformed metal or silicon oxide preformed material, instead of gelation of the inorganic oxide network in

the presence of indifferent chemicals resulting in the formation of an interpenetrating inorganic/organic network, as in Novak.

For the forgoing reasons, it is respectfully submitted that the rejection of Claims 23, 25-31, 36, 39-42, 47-49, 69, 70 and 71 under 35 U.S.C. 102(b) is inapplicable to the Claims as amended and its withdrawal is respectfully requested.

The Examiner has also rejected Claims 23, 25-31, 36, 39-42, 47-49, 69, 70 and 71 under 35 U.S.C. 102(b) as assertedly being anticipated by Yim et al. Korean J. Chem. Eng. 2002, 19, 159-166. The Examiner contends that "Yim et al. disclose preparations of polyisocyanate cross-linked aerogels prepared by forming a sol-gel material as defined by applicants' claims, solvent to form a wet-gel film followed by drying...." The Examiner further contends that "applicants' arguments ... do not identify differences over Yim et al. which are supported by corresponding limitations in the claims." To the extent the foregoing rejection can not be applied to the Claims as amended, this ground of rejection is respectfully traversed.

As pointed out in the discussion above with respect to Novak, in the present invention, a three step process is used. The first step of the present invention, which is absent in Yim, is "forming a metal oxide or silicon oxide based sol-gel preformed material prior to reaction with organic cross-linking agents." (Emphasis added.) Then, the preformed metal oxide or silicon oxide based sol-gel perform material is contacted with "an organic cross-linking agent, the cross-linking agent comprising an organic compound." (New language added.). This results in an "organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material," as specifically recited in Claim 23. (Claim 25 includes a similar limitation.) The last step is "drying the cross-linked metal oxide or silicon oxide based sol-gel preformed material provided with a conformal coating of chemically bound organic

material to form the porous cross-linked metal oxide or silicon oxide based aerogel material.”

(New language underlined.)

Yim employs a different procedure and obtains a different material. Yim’s process is as follows: “First, tetramethyl orthosilicate (TMOS), water, and HCl were added into a beaker by the molal ratio of 1:1.3:10⁻⁵. Then, this feed solution was mixed by stirrer for an hour. Second, produced alcohol was removed from this feed solution by a batch distillation. Third, the feed sol solution was partially hydrolyzed to obtain condensed silica solution. [Citation omitted]. In this partially condensed solution (CS), polymeric MDI solution was added by varying density and the amount of catalyst to reach silica organic hybrid aerogel.” (Yim, at 161, col. 1, lines 35-9).

(Emphasis added).

Because Yim adds the polymeric MDI solution to a “partially” condensed silica solution, gelation of the silica and reaction of the silica particles with isocyanate both proceed simultaneously. Thus, Yim does not “perform[] a metal oxide or silicon oxide based sol-gel preformed material prior to substantial cross-linking by organic compounds,” as specifically recited in Claim 23 (and similarly in Claim 25). Yim’s simultaneous silica gelation and organic polymerization procedure causes a very long gelation time of about a week. (Yim, at 161, col. 2, lines 15-16).

One skilled in the art would recognize that the reason Yim’s gelation takes a very long time is because polymeric MDI reacts with the surface silanol groups of partially condensed silica, therefore interfering with gelation of the silica. This interference also disrupts the formation of the network of metal oxide or silicon oxide particles, causing the formation of a different material from the material formed by Applicants’ process. Although somewhere Yim’s process does result in formation of a gel that incorporates isocyanate-derived polymer, nowhere

does Yim teach or suggest that his different process results in a material comprising “organic conformal coating chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material,” as specifically recited in Claim 23. (Claim 25 includes a similar limitation.).

For the forgoing reasons, it is respectfully submitted that the rejection of Claims 23 and 25 under 35 U.S.C. 102(b) is inapplicable to the Claims as amended and its withdrawal is respectfully requested.

Claims 31, 36, 39-42, 47-49, 69, 70 and 71 are dependent upon one or the other of Claims 23 and 25 and include additional limitations.

Furthermore, with respect to Claim 39, the Examiner contends that the claimed degree of rupture resistance would be “inherent” in the preparations of Yim. Applicants respectfully disagree. “Inherency” does not exist where a result might be the case, “inherency” exists only where no other result is possible. Here, because Yim does not employ the same process as Applicants’ claims, it cannot be concluded a priori that it is “inherent” that Yim’s materials will have the same properties. Furthermore, as has been demonstrated in Applicants’ prior responses to earlier Official Actions, Yim’s material does not have the same properties as Applicants’ material. Accordingly, withdrawal of the assertion of inherency, and withdrawal of the rejection of Claim 39 on that basis, are relieved in order and courteously requested.

Claim Rejections - 35 USC §103 – Claims 26-31

The Examiner has rejected Claims 26-31 under 35 U.S.C. 103(a) over Yim and Novak on the asserted basis that Applicants’ claimed materials are different from Yim’s and Novak’s only in the differences in the amounts of the crosslinking component. This rejection is respectfully traversed. In fact, the materials obtained by the three-step process of Claim 23 (and Claim 25)

are fundamentally different from the materials obtained by the processes taught in either Yim or Novak.

The Examiner contends that Applicants' materials are identical to Yim's and that his materials only "differ from applicants' claims in that he does not specifically specify amounts of the polyisocyanate group containing component that are used in their preparation [and that] normally, changes in result effective variables are not patentable where the difference involved is one of degree, not a kind." This contentions is respectfully submitted to be erroneous. As discussed above with respect to the Examiner's rejections under 35 U.S.C. 102(b), the materials obtained by the three-step process of Claim 23 are fundamentally different from those obtained by Yim's mixing of his isocyanate with the sol-gel precursors while the sol-gel precursors are only "partially" condensed.

The Yim article does not show structural characterization of the material produced by his method. However, in a review article on the subject relied upon by experts in this field as a learned treatise (including the review article by Husing et al. *Angew. Chem. Int. Ed.* 1998, 37, 22-45, which is of record), describing material formed by the same protocol that Yim later described:

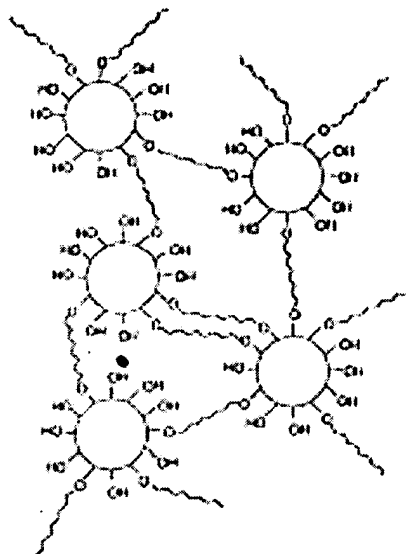


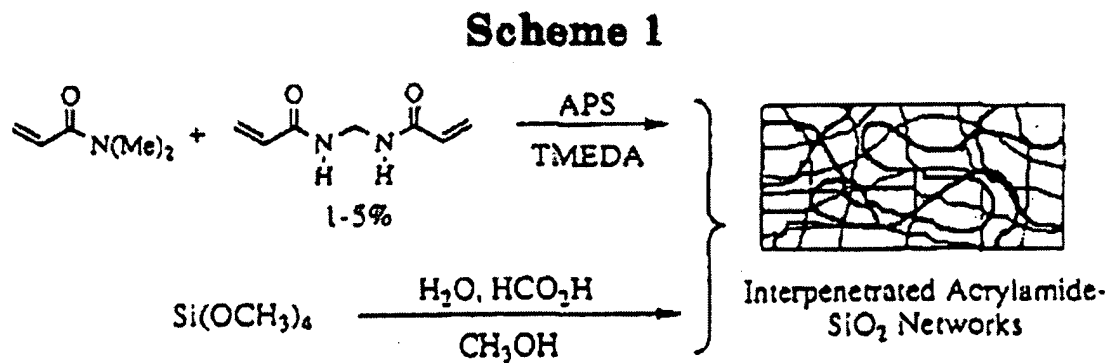
Figure 8 Schematic representation of an alumina gel cross-linked with hexamethylenediisocyanate (reproduced with friendly permission from ref. [11]). The big circles represent particles of hydroxylated alumina, and the wiggly lines $\text{O}(\text{NH}(\text{CH}_2)_6\text{NH}(\text{O}))$ chains.

The above-diagram shows one way in which Yim's materials are different from Applicants' claimed materials. In this diagram, the wiggly lines show polymer chains formed between silicon oxide particles, the polymer chains being formed prior to performing of the network of silica particles that occurs during gelation. By contrast, in Applicants' invention, by *performing* the silica (or metal oxide) sol-gel network, Applicants ensure that the isocyanate (the wiggly lines in the scheme above) do not interfere with gelation of the silica. By introducing the isocyanate in a second step (see claim 23), *after the silica particle network has been formed*, we ensure that: (a) the isocyanate reacts with the exposed surfaces of the nanoparticles; (b) coats the particle network conformally; and (c) it does not separate silica particles (as in Yim) because the particle network has already been formed.

The distinction between the material formed by Applicants' process from the material formed by Yim's process is also shown by the SEM pictures of Applicants' specifications

(Applicants' Figs 2A and 2B). These SEM pictures show that, although there has been a heavy uptake of crosslinker (up to 70% w/w) the structure of the silica nanoparticle network has not been affected or changed. That is, because Applicants' silica nanoparticle network is preformed prior to reaction of the cross-linking agent, crosslinking by the organic polymer results in a conformal coating on the surface of the metal oxide or silicon oxide preformed material, but does not change the structure of the metal oxide or silicon oxide preformed material.

Applicants' claimed materials are also fundamentally different from Novak's. Novak's process of mixing sol-gel precursors with polymers and/or with precursors that form polymers that do not interact chemically with the sol-gel network, resulting is a different material. Novak himself shows that his process results in an *interpenetrated* network of polymer chains in the voids of the silica network of particles. See, as depicted in Novak's "Scheme 1" figure. (Novak, page 284, col. 1, "Scheme 1").



Furthermore, according to the review article by Husing et al. Angew. Chem. Int. Ed. 1998, 37, 22-45 as discussed above, which article is cited by relied upon by experts in this field as a learned treatise, all Novak's protocols yield materials which are invariably are referred to in

the art as *interpenetrating organic networks*. Please refer to Figures 11 and 12 of the Husing review article, referring directly to Novak's paper cited by the reviewer:

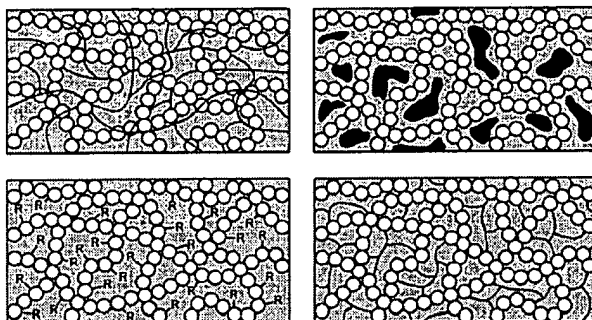


Figure 11. Inorganic-organic hybrid networks obtained by sol-gel processing. Top left: interpenetrating inorganic and organic networks; top right: incorporation of organic molecules (enzymes, dyes, etc.); bottom left: modification (functionalization) of oxidic materials with organic substituents; bottom right: dual networks.

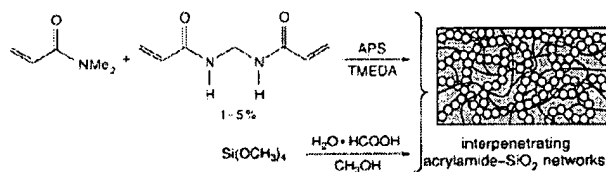


Figure 12. Formation of two interpenetrating networks (inorganic-organic) by simultaneous sol-gel reaction and polymerization of *N,N*-dimethylacrylamide (in the presence of *N,N'*-methylenebisacrylamide (APS = ammonium persulfate); according to ref.[132]).

By contrast to the *interpenetrating* organic network formed by Novak's process, in Applicants' invention defined by Claim 23 the different process results in "an *organic conformal coating* chemically bound to surfaces of said metal oxide or silicon oxide based sol-gel preformed material." (Emphasis added). Claim 25 includes a similar limitation, "a *conformal coating* of an organic substance formed by *chemical bonding* of an organic cross-linking agent to surfaces of said metal oxide or silicon oxide based sol-gel preformed material after formation of said metal oxide or silicon oxide based sol-gel preformed material." (Emphasis added.) Accordingly, both Claims 23 and 25 clearly distinguish from Novak's "interpenetrating organic networks." Furthermore, as pointed out in response to prior Official Actions, the materials of Applicants' invention have significantly different properties than the materials described in Novak and Yim, further

demonstrating that the materials defined by Claims 23 and 25 are fundamentally different materials..

For the foregoing reasons, as well as the reasons previously expressed in previous responses, that the new materials defined by the limitations of Applicants' Claims 23 and 25 would not have been obvious in view of Novak and Yim,.

For the foregoing reasons, it is respectfully submitted that the inventions defined by Claims 23, 25, 26-31, 36, 39-42, 47-49, 69, 70 and 71 would not have been obvious, even apart from the differences in the amounts of crosslinker uses, as defined in Claims 26-31. Withdrawal of the rejection of these claims under 35 U.S.C. 103 (a) is believed is believed in order and courteously solicited.

New Claim 72 has been added. Claim 72 is similar to Claim 25, but also includes the limitation "wherein the volume relaxation energy (VRE) of the cross-linked metal oxide or silicon oxide based sol-gel material upon evaporation of the pore-filling solvent is less than the energy required to destroy the chemical bonding associated with cross-linking." Claim 72 is submitted to be allowable for all of the reasons stated above with respect to Claim 25, as well as by reason of its additional limitation.

For the foregoing reasons, it is respectfully submitted that the invention defined by Claims 23, 25-31, 36-42, 45-49, 65 and 69-72 is neither anticipated by, nor obvious in view of the prior art of record. Accordingly withdrawal of the rejections of Claims 23, 25-31, 36-42, 45-49, 65 and 69-71 and the allowance of new Claim 72 claims under 35 U.S.C. 102(b) and/or 103 (a) and full allowance of Claims 23, 25-31, 36-42, 45-49, 65 and 69-72 are believed in order and is courteously solicited.

Please charge the fee of \$105.00 for a new independent Claim and \$60.00 for an extension of time of one month for a small entity, and any other additional fees incurred by reason of this

amendment, to our Deposit Account No. 50-0605 of CARR LLP.

Respectfully submitted,

CARR LLP

Date: November 17, 2008

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